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INK RECEPTIVE ELEMENT AND PRINTING METHOD  
[Inku Juri Yoso Oyobi Insatsu Hoho]

Mizue Fukushima, et al.

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INVENTORS	(72): FUKUSHIMA, MIZUE; TAKIZAWA, MAKI
APPLICANT	(71): CITIZEN WATCH CO., LTD.
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Title

Ink Receptive Element and Printing Method

Technical Field

The present invention pertains to an ink receptive element. More specifically, it pertains to an ink receptive element on which printing can be implemented with an aqueous ink by an ink jet method, most of all, to an ink receptive element that has a base material that in itself has no ink receptivity—for example, plastics, glass, ceramics, metals, etc. The ink receptive element of the present invention can be used as, for example, clockfaces; the dial plates, operating panels, etc., of measuring instruments; and the dial plates of speedometers for automobiles, motorcycles, etc. The present invention also pertains to a method for implementing printing on this ink receptive element. In the present specification, the term "inji ["printing" in Japanese]" is also used in the same sense as "insatsu ["printing" in Japanese]."

Prior Art

It is a well-known fact that ink jet methods are currently widely used as printing techniques or recording techniques. For example, for office automation equipment, such as word processors, personal computers, facsimile machines, etc., and various kinds of measuring instruments, such as measuring instruments for medical use, printers

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that utilize ink jet methods, that is, ink jet printers, are widely used in order to print data output from this type of equipment on recording paper. Ink jet printers have many advantages—for example, because regular paper can be used as recording paper, their running cost is low; they do not require additional process steps, such as transfer, fusion, etc.; and they can be easily configured for color printing and are capable of producing sharp color prints.

Printing with ink jet printers is usually carried out on paper, such as regular paper, etc., considering the properties of inks used /2 for printing, the mechanisms of printing, and so forth. In recent years, however, ink jet printing is also carried out on recording mediums other than paper (in the present specification, they are referred to as "ink receptive elements")—for example, sheets, plates, etc., comprised of such hard materials as metals, glass, plastics, etc.

However, hard materials, such as those cited in the foregoing, do not wet well with ink jet inks and, consequently, do not allow the inks to firmly adhere to them, and, accordingly, various attempts have been made to solve this problem. In JP-B-S55-034712, for example, is disclosed an ink jet recording method that uses a water-soluble ink and according to which a water-soluble coating material is applied to a medium to be recorded beforehand, and ink jet recording is carried out before the coated surface sets to touch. That is to say, this method sprays a water-soluble ink while there is still enough moisture remaining in the coating material and thus utilizes the bleeding

phenomenon of the ink in order to improve the thickness of recorded images and the wettability of the ink particles.

In JP-B-S55-034713 is disclosed a jet printing method according to which, after an emulsion-type coating material whose pigment volume concentration is 20% or higher is applied to a base material having no absorbency and dried, non-drying ink particles are injected at a high speed continuously and stably through a thin nozzle having an inner diameter of approximately 10 to 400  $\mu\text{m}$ . That is to say, this method forms a porous film having a high pigment concentration and thereby attempts to make it easier for the ink to disperse on the surface of the film and inside it.

In JP-B-S60-027589 is disclosed a method for forming images on plastic sheets by an ink jet technique, said method being characterized by the fact that a sublimating dye is used as part or all of the components of the coloring matter of the ink, while a resin whose curing progresses with ultraviolet rays is used as a dispersion medium, and, after the image formation, ultraviolet irradiation of the sheet and the increase of temperature of the sheet by heating are simultaneously or consecutively carried out to achieve fusion. This /3 method uses a sublimating dye for the purpose of improving the permeability of the ink into plastic sheets and also uses an ultraviolet curing resin as the dispersion medium of the aforesaid dye so as to implement ultraviolet irradiation and thermal fusion in

combination for the purpose of imparting wear resistance to plastic sheets.

In JP-A-H09-314986 is disclosed an aluminum plate for ink jet recording use, said aluminum plate being characterized by the fact that the aluminum base plate has an anodic oxide film on the surface and that on this film is further formed an ink receptive layer having excellent ink absorbency. The ink receptive layer is formed from a water-soluble polymer, such as a polyvinyl alcohol, acrylic acid ester polymer, carboxy methyl cellulose, starch, casein, etc., in a thickness of 7 to 15  $\mu\text{m}$ . The aluminum plate of this invention has an anodic oxide film formed between the aluminum base plate and the ink receptive layer so as to improve the adhesion strength of the ink receptive layer to the base plate and also to prevent the deterioration, such as rusting, etc., of the base plate.

To manufacture various kinds of clockfaces and the like, it is a common practice to print desired characters, symbols, etc., on the surface of an undercolor-coated metal plate or plastic plate with oil-based ink by bat printing, screen printing, etc. This method, however, is troublesome and requires high production cost, and, therefore, it is desirable to employ ink jet methods, such as those described in the foregoing, for printing patterns, characters, symbols, etc., because ink jet methods, as explained in the foregoing, have advantages in, for example, that they are easily adapted to color printing and can produce sharp color prints.

To print patterns, etc., by an ink jet method in the production of dial plates, etc., several requirements must be met. A first requirement is that, because a base material, such as a metal plate, plastic plate, etc., that in itself has no ink receptivity is used, /4 an aqueous ink suitable for ink jet methods must be received completely by its surface and also be firmly attached to it, in other words, the ink ejected toward the base material should adhere to its surface, should not run on its surface, and should not be readily wiped off once dried.

To meet this first requirement, it is conceivable to provide an ink receptive layer on the base material. However, the ink receptive layer must also satisfy the following important requirements.

1. It must be transparent.

Because the texture of the base is an important factor for the improvement of design, etc., the ink receptive layer must have a sufficient degree of transparency that allows the coated surface of the clockface and metallic luster to show through. Consequently, although it is normally preferable to form an ink receptive layer by dispersing porous filler particles in a transparent binder resin for the purpose of improving the ink absorption speed and ink absorption quantity of the ink receptive layer, this method cannot be employed here because it inhibits transparency.

2. It must absorb and fix ink well so as not to cause bleeding or color mixing.

If this requirement is not met, high print quality cannot be obtained, and the decrease of process yield cannot be avoided.

3. It must have excellent environmental resistance.

Since the dial plates of clocks, etc., are often exposed to severe environments, the ink receptive layer must have a high level of environmental resistance. For example, it must at least pass the following environmental tests. Each environmental test will be described in detail later.

(1) Moisture resistance test---40 °C/90% RH (relative humidity), 100 hours

(2) Lightfastness test (dry and wet)--- Sunshine Weatherometer, 100 hours

(3) Adhesion test (cross-cut test)--- Pursuant to JIS D0202 8.12 /5

However, to the best of the present inventors' knowledge, no ink jet printing method that can meet the aforesaid requirements when printing patterns, etc., of dial plates, etc., has been proposed yet, and neither has an ink receptive element that is suitable for this type of printing method been proposed yet. The ink jet printing methods that were cited and explained in the foregoing cannot meet these requirements, either.

For example, the method proposed in JP-B-S55-034712 applies a water-soluble coating material to a recording medium beforehand to form a coating corresponding to the ink receptive layer of the present invention, but, since this coating is colored, it does not satisfy the



requirements, such as transparency, etc., of the ink receptive layer and also denies the presence of the undercolor coating, which could contribute to the improvement of design, etc. In addition, since this method requires a special process step in which ink jet recording is carried out before the surface of the coating sets to touch, it is unavoidable for the recording process to become troublesome and the equipment to become complex.

The method proposed in JP-B-S55-034713 also has similar problems. The reason for this is that this method applies and dries a coating material having a high pigment volume concentration prior to the printing of dial plates, etc., and the colored coating obtained in this manner corresponds to the ink receptive layer of the present invention.

With the image forming method disclosed in JP-B-S60-027589, since there is no transparent coating corresponding to the ink receptive layer of the present invention, the effects derived from it cannot be expected. In addition, since this method limits the base material to plastic sheets, it lacks versatility.

Similarly, the aluminum plate for ink jet recording use disclosed in JP-A-H09-314986 also lacks versatility in that the base material is limited to an aluminum plate. In addition to this shortcoming, the /6 ink receptive layer employed here is comprised of a quite common water-soluble polymer, and it can only yield the effect of improving ink absorbency and cannot satisfy such requirements as high print

quality, transparency, environmental resistance, etc. Furthermore, because it is necessary to place an anodic oxide film between the aluminum plate and the ink receptive layer, the effect of the undercolor coating is limited to the effect of the color derived from the anodic oxide film, and, consequently, this method cannot yield the texture effect that the present inventors hope to achieve.

#### Disclosure of the Invention

One object of the present invention is to provide an ink receptive element that has a base material that in itself does not have ink receptivity, said ink receptive element being particularly suitable for ink jet methods.

Another object of the present invention is to provide an ink receptive element whose ink absorption speed and absorption quantity can be improved to a degree comparable or superior to an ink receptive layer formed by dispersing porous filler particles in a transparent binder resin, without requiring an ink receptive layer thus formed.

Yet another object of the present invention is to provide an ink receptive element that, if there is an undercolor coating as the base, allows its coated surface or metal luster to show through, that can realize high print quality, and that has excellent environmental resistance.

Yet another object of the present invention is to provide an ink receptive element that, in the process of forming an ink receptive

layer on the base material, exhibits high adhesion strength of the ink receptive layer to the base material.

Yet another object of the present invention is to provide a method for implementing high quality printing by ink jet techniques on an ink receptive element that has a base material that in itself does not have ink receptivity.

These and other objects of the present invention will become apparent from the following description. /7

The present invention, in one aspect, provides an ink receptive element containing a base material that in itself does not have ink receptivity, the aforesaid ink receptive element having, on its printing surface, a transparent ink receptive layer that contains, as the main component, a film-forming polymer prepared by the polymerization of a monomer that is soluble in water and that has hydrophilic moieties in its molecules.

The present invention, in another aspect, provides a method for implementing printing on an ink receptive element containing a base material that in itself does not have ink receptivity, said method being comprised of forming, on the printing surface of the aforesaid ink receptive element, a transparent ink receptive layer that contains, as the main component, a film-forming polymer prepared by the polymerization of a monomer that is soluble in water and that has hydrophilic moieties in its molecules and subsequently of performing printing with a water-based ink by an ink jet method.

## Brief Explanation of the Drawings

Fig. 1 is a sectional drawing illustrating a preferable embodiment of the ink receptive element pertaining to the present invention.

Fig. 2 is a sectional drawing illustrating another preferable embodiment of the ink receptive element pertaining to the present invention.

Fig. 3 is a sectional drawing illustrating another preferable embodiment of the ink receptive element pertaining to the present invention.

Fig. 4 is a sectional drawing illustrating yet another preferable embodiment of the ink receptive element pertaining to the present invention.

Fig. 5 is a schematic drawing illustrating one example structure of the film-forming polymer contained in the ink receptive layer of the ink receptive element of the present invention.

Fig. 6 is a chemical formula representing concrete examples of the film-forming polymer shown in Fig. 5.

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Fig. 7 is a schematic drawing illustrating another example structure of the film-forming polymer contained in the ink receptive layer of the ink receptive element of the present invention.

Fig. 8 is a chemical formula representing concrete examples of the crosslinking monomer used to form the film-forming polymer shown in Fig. 7.

Fig. 9 is a schematic drawing illustrating the pre- and post-polymerization behavior of a light-curing polymer that is concomitantly used for the preparation of the film-forming polymer.

Fig. 10 consists of sectional drawings that sequentially illustrate a preferable embodiment of the ink jet printing method of the present invention.

Fig. 11 is a schematic drawing illustrating the method for measuring dynamic contact angles used for the evaluation of hydrophilicity.

Fig. 12 is a graph obtained by plotting the relationship between the duration of the ultraviolet-ozone treatment and the dynamic contact angle.

Fig. 13 is a graph obtained by plotting the relationship between the duration of the ultraviolet-ozone treatment and color difference  $\Delta E^*_{ab}$ .

#### The Best Mode of Implementing the Invention

The ink receptive element pertaining to the present invention consists, at a minimum, of a base material and an ink receptive layer provided on the printing surface (on which a desired print is formed) of the base material. The ink receptive layer is normally provided only on one side of the base material, but, if necessary, it may be provided on both sides of the base material.

Fig. 1 illustrates a typical example of the ink receptive element of the present invention. The ink receptive element (10) has an ink

receptive layer (2) provided directly on one side of its base material (1). On the ink receptive layer (2) are sprayed ink droplets (9) that are ejected from an ink jet printer (not shown). Here, the base material (1) may be formed from any kind of material and in any mode, /9 but, as can be seen from the previous explanation, materials, such as paper or recording paper similar to it, that have sufficient ink receptivity in themselves are excluded from the scope of the present invention. The example shown in the figure has a base material comprised of a plastic material.

Suitable materials used as the base material (1) for implementing the present invention include plastic materials, such as polyester resins, polyolefin resins, polyvinyl chloride, acrylic resins, phenolic resins, etc.; glasses, such as oxide glasses, silicate glasses, etc.; ceramics; metals, such as aluminum, steel, copper, titanium, etc.; wood; and the like. However, the base material used for implementing the present invention may be any materials other than those listed above as long as they do not have ink receptivity in themselves and do not adversely affect the effects of the present invention.

With respect to the shape of the base material (1), besides a sheet shape, as shown in Fig. 1, it may be in the shape of plate, block, sphere, cylinder, etc., and its printing surface may have an embossed pattern formed on it. The size of the base material (1) can also be changed optionally according to the target printed products.

The ink receptive layer (2) must be above all transparent. The reason for this is that, if it were translucent or opaque or colored, the texture of the base could not be utilized to improve design, etc. Taking a clockface as an example, it has a metal plate and an undercolor coating (coating film) provided on the plate, on which patterns, characters, symbols, etc., will be further printed, and, if the ink receptive layer is not transparent here, the metal luster or undercolor-coated surface cannot be utilized to improve the texture. Consequently, the ink receptive layer (2) used in the present invention cannot contain fillers, such as porous inorganic particles, etc., that are conventionally mixed into ink receptive layers to improve their ink absorption speed and absorption quantity according to the prior art. The ink receptive layer (2), of course, may /10 optionally contain a proper amount of additives, such as coating aids, etc., within a range that does not adversely affect the transparency of the ink receptive layer. The degree of transparency of the ink receptive layer (2), when measured as the transmittance of light having a wavelength of 500 nm (measured with a spectrophotometer), is usually preferably 90% or higher, better yet, 95% or higher.

In addition, it is essential for the ink receptive layer (2) to contain, as its main component, a film-forming polymer that is formed by the polymerization of a monomer that is soluble in water and that has hydrophilic moieties in its molecules (which may hereinafter be referred to as "a water-soluble monomer"). The present invention

configures the ink receptive layer from a film-forming polymer having this type of specific structure, thereby obtaining various effects, including the effect of making it possible to form a transparent film, the effect of increasing the affinity of the ink receptive layer for ink and thereby making it possible to absorb a large quantity of ink rapidly, the effect of preventing color bleeding and color mixing, the effect of imparting high environmental resistance to the ink receptive layer, and so forth, without mixing fillers or other additives or carrying out other treatments, which is the case with the prior-art ink receptive layers. The film-forming polymer can usually be produced by formulating a film-forming composition containing a water-soluble monomer and other essential components and subsequently by polymerizing the aforesaid water-soluble monomer in this film-forming composition. The ink receptive layer can be formed by applying a resin solution containing this film-forming polymer onto the base material and drying it.

Fig. 5 is a schematic drawing illustrating one example structure of the film-forming polymer that is contained in the ink receptive layer and that is derived from the aforesaid water-soluble monomer. As shown in the figure, the hydrophilic moieties (A) are contained in parts of the main chains (L) of the film-forming polymer. In the example illustrated in the figure, the hydrophilic moieties (A) bond with parts of the main chain (L) of the film-forming polymer, but, as



long as the desired operational effects can be obtained, the bonding mode and quantity of the hydrophilic moieties (A) can be varied optionally. Fig. 6 is the chemical formula of a typical example of /11 the film-forming polymer illustrated in Fig. 5, that is, a polymer formulated from N,N-dimethyl acrylamide as the water-soluble monomer. In the formula, n represents the number of the repeating units of the N,N-dimethyl acrylamide monomer, and Me represents a methyl group. With this polymer, each NC=O can function as a hydrophilic moiety (A).

The water-soluble monomer used for the formulation of the film-forming polymer is not limited in any specific way as long as it meets the given conditions, but the hydrophilic moiety to be contained in its molecule is preferably an amide group, as mentioned in the foregoing. Accordingly, water-soluble monomers suitable for implementing the present invention include, but are not limited to, substituted or non-substituted acrylamide compounds, such as acrylamide, N-methyl acrylamide, N-ethyl acrylamide, N,N-dimethyl acrylamide, N,N-diethyl acrylamide, etc.; substituted or non-substituted methacrylamide compounds, such as methacrylamide, N-methyl methacrylamide, N-ethyl methacrylamide, N,N-dimethyl methacrylamide, N,N-diethyl methacrylamide, etc.; acryloyl morpholine, etc.; and such compounds as diacetone acrylamide, N-isopropyl acrylamide, etc., which can become water soluble by copolymerizing with other water-soluble monomers. Of these water-soluble monomers, N,N-dimethyl acrylamide is

particularly suitable from the standpoint of transparent-film-forming ability and print quality.

The thickness of the ink receptive layer (2) configured from the film-forming polymer derived from a water-soluble monomer is not limited in any specific way and can be varied in a wide range according to the desired effects, the quantity of ink to be accepted by it, and so forth. Usually, the thickness of the ink receptive layer (2) is preferably in the range of several to 50  $\mu\text{m}$ , better yet, in the range of 5 to 30  $\mu\text{m}$ . If the ink receptive layer (2) needs to accept a /12 relatively large amount of ink, its thickness may exceed 50  $\mu\text{m}$ .

The process of polymerizing a monomer to formulate a film-forming polymer corresponding to it can be implemented, as a rule, according to a conventional polymerization method suitable for each individual monomer. For example, the polymerization can be implemented by reacting a given quantity of a selected water-soluble monomer in the presence of a polymerization initiator in a nitrogen atmosphere at a temperature increased to 50 to 70 °C. The polymerization initiator used here is preferably a water-soluble polymerization initiator. Suitable polymerization initiators include 2,2'-azobis (2-amidinopropane) dihydrochloride, 4,4'-azobis (4,4-cyanovaleric acid), 2,2'-azobis [2-(2-imidazoline-2-yl) propane] dihydrochloride, 2,2'-azobis [2-(imidazoline-2-yl) propane] disulfate dihydrate, 2,2'-azobis isobutyl amide dihydrate, persulfate, etc. In addition, polymerization

accelerators and other additives may be added to the film-forming composition, as necessary. Since the polymerization conditions of this type of water-soluble monomer can be found in many technical documents of the field of the polymer science, please refer to these technical documents for specifics.

In addition to the layer structure that was explained in reference to Fig. 1, the ink receptive element of the present invention may assume various layer structures within the scope of the present invention. Figs. 2 through 4 each illustrate another preferable embodiment of the ink receptive element of the present invention. Furthermore, although not illustrated, the ink receptive element of the present invention may additionally have, as necessary, other layers, such as a light-reflecting layer, overcoat layer, etc., that are conventionally provided for ink receptive elements

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The ink receptive element (10) illustrated in Fig. 2 has an undercolor coating (3) between the plastic base material (1) and the ink receptive layer (2). By placing this kind of colored undercolor coating (3) beneath the transparent ink receptive layer (2), texture improvement derived from the undercolor coating (3) can be achieved. If, for example, the undercolor coating (3) is comprised of a metallic coating or pearl mica coating, its metallic luster can be observed through the transparent receptive layer (2). In addition, the design effect of causing subsequently printed characters, symbols, etc., to stand out in the metallic luster can be achieved.

The undercolor coating (3) can be readily formed using conventional techniques. For example, it can be formed by applying a solution or powder comprised of dyes, pigments, solvents, etc., onto the base material by coating, painting, etc., in a desired thickness and pattern and by curing it. The coating can be implemented, for example, by roll coating, brush coating, etc., and the painting can be implemented by electrophoretic painting, electrostatic painting, etc. In place of the undercolor coating, a metallized film or anodic oxide film may be formed, as necessary.

The thickness of the undercolor coating (3) can be varied in a wide range according to the type of the ink receptive element and so forth, but it is usually preferable to set it in the range of 10 to 200  $\mu\text{m}$ , better yet, in the range of 20 to 100  $\mu\text{m}$ . The thickness of this undercolor coating (3) can, however, be outside the aforesaid range, depending on the type of the employed film-forming method.

The ink receptive element (10) illustrated in Fig. 3 has an undercolor coating (3) between the plastic base material (1) and the ink receptive layer (2), as the ink receptive element (10) shown in Fig. 2, and also has a clear coating (4) formed on the surface of the undercolor coating (3). The clear coating (4) has, for example, the effect of further improving the color effect of its base, that is, the undercolor coating (3). The clear coating can be formed by applying a clear coating material of the urethane type, acrylic type, epoxy

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type, etc., and by curing it. In addition, in place of a clear coating material, a white coating material, etc., may be used as necessary.

For the ink receptive element of the present invention, after the undercolor coating (3) is formed in the aforesaid manner or after the undercolor coating (3) is formed and, subsequently, the clear coating (4) is further formed, it is preferable to perform a hydrophilicity-imparting treatment so as to suppress the hydrophobicity of these coatings. The hydrophilicity-imparting treatment here can be any conventional hydrophilicity-imparting treatment, such as an ultraviolet-ozone treatment, a treatment using a coupling agent, an etching treatment, etc. Of these, an ultraviolet-ozone treatment is especially effective. Since this treatment imparts high hydrophilicity to the surface of the ink receptive element that is still in the middle of its production process, the ink receptive layer can be attached to its surface with higher adhesion strength.

The ink receptive element (10) illustrated in Fig. 4 is an example that has a base material (11) made from a metal in place of the plastic base material. For the example illustrated in the figure, a base material (11) made of brass that has anti-corrosion plating (12) around it is used. On this base material (11) are formed, as the ink receptive element (10) shown in Fig. 2, an undercolor coating (3) and subsequently an ink receptive layer (2). Although not illustrated, it is preferable for the ink receptive element (10) to have a clear

coating that has been subjected to a hydrophilicity-imparting treatment as the base for the ink receptive layer (2).

The ink receptive layer, which plays the most important role in the ink receptive element of the present invention, contains, as its main component, a film-forming polymer that is produced by the polymerization of a monomer (water-soluble monomer) that is soluble in water and that has hydrophilic moieties in its molecules. This film-forming polymer is, as described in the foregoing, typically a homopolymer obtained by the polymerization of a water-soluble monomer, but, if so desired, it may be a copolymer of this water-soluble monomer and another water-soluble monomer. The use of this film-forming polymer in the form of a copolymer makes it possible to /15 achieve additional effects, such as improvement of moisture resistance, improvement of water resistance, prevention of print-quality degradation, and the like. In addition, the film-forming polymer of the present invention may be in the form of a mixture comprised of it and other polymers.

According to a preferred mode of the present invention, the film-forming polymers that can assume the form of a copolymer or a mixture include, but are not limited to:

1. a copolymer produced by the polymerization of a water-soluble monomer and a crosslinking monomer that is soluble in water and that has hydrophilic groups in its molecules,

2. a mixture comprised of a homopolymer derived from a water-soluble monomer and of a light-curing polymer having hydrophilic groups and photopolymerizable functional groups in its molecules, and
3. a mixture comprised of a copolymer produced by the polymerization of a water-soluble monomer and a crosslinking monomer that is soluble in water and that has hydrophilic groups in its molecules and of a light-curing polymer having hydrophilic groups and photopolymerizable functional groups in its molecules.

The copolymer described in the aforesaid item 1 (hereinafter referred to as "Copolymer 1") is a copolymer produced by the polymerization of a water-soluble monomer and a crosslinking monomer. The crosslinking monomer used here is required not only to be soluble in water but also to have a plurality of crosslinking points in its molecule. There should be a minimum of two crosslinking points in its molecule.

Crosslinking monomers that are suitable for the formation of Copolymer 1 and that have two or more crosslinking points in their molecules include, but are not limited to, acrylamide-based or methacrylamide-based crosslinking monomers, such as methylene bisacrylamide, methylene bismethacrylamide, bisacrylamide acetic acid, N-methylol acrylamide, N-methylol methacrylamide, N,N'-(1,2-dihydroxy) ethylene bisacrylamide, triacryl formal, etc. In place of these crosslinking monomers may be used water-soluble acrylic ester or methacrylic ester monomers that have two or more crosslinking points

and also have a plurality of ethylene glycol chains and (or) propylene glycol chains in a molecule, some examples of such monomers including diacrylic ester, dimethacrylic ester, triacrylic ester, trimethacrylic ester, etc.

The polymerization for forming Copolymer 1, as the formation of homopolymers explained before, can be implemented by conventional polymerization methods under commonly employed polymerization conditions.

Fig. 7 is a schematic drawing illustrating one example structure of Copolymer 1 (film-forming copolymer) derived from the copolymerization of the aforesaid water-soluble monomer and the aforesaid crosslinking monomer. As can be seen from the figure, this copolymer has a three-dimensional structure, unlike the homopolymer having the linear structure shown in Fig. 5. More specifically, in addition to the bonding of the hydrophilic moieties (A) to the main chains (L) of the copolymer, the copolymerization of the water-soluble monomer and the crosslinking monomer causes the crosslinking monomer having hydrophilic groups (B) to bridge between the main chains (L), thereby creating a crosslinked structure. Fig. 8 is a chemical formula of polyethylene glycol diacrylate that is useful as the crosslinking monomer in the formation of Copolymer 1. Introduction of this type of crosslinking monomer is effective for imparting water resistance as well as for preventing the degradation of print quality. In particular, those that have an ethylene glycol chain, that is, the hydrophilic



group (B) in the formula shown in Fig. 8, in a molecule have excellent affinity with pigment-based aqueous ink and, as such, are useful for achieving improved print quality.

The polymer mixture described in the aforesaid item 2 (hereinafter referred to as "Mixture 2") is a mixture prepared by forming a film-forming polymer by polymerizing a water-soluble monomer and subsequently by mixing it with a photopolymerizable (light-curing) polymer that is soluble in water and that has hydrophilic groups and photopolymerizable functional groups in its molecules. The polymer mixture described in the aforesaid item 3 (hereinafter referred to as "Mixture 3") is a mixture prepared by forming Copolymer 1 mentioned above by polymerizing the aforesaid water-soluble monomer and the aforesaid crosslinking monomer and subsequently by mixing it with a light-curing polymer that is soluble in water and that has hydrophilic groups and photopolymerizable functional groups in its molecules. With respect to these polymer mixtures, the use of light-curing polymers having a specific structure that is soluble in water before photopolymerization and becomes insoluble in water (that is, hydrophobic) after photopolymerization makes it possible to further improve moisture resistance and water resistance. /17

Preferable light-curing polymers that can be used for the formation of both Mixtures 2 and 3 are ultraviolet-curing polymers. The curing of these polymers by irradiation of ultraviolet rays is

preferably implemented not immediately after the formation of the ink receptive layer by application and drying of a resin solution containing these polymers on a base material but after required printing is performed on the ink receptive layer thus formed. The reason for this is that print quality is adversely affected if the curing is implemented prior to the printing process. On the other hand, if the curing is implemented after the printing process is completed, the ink is incorporated into the ink receptive layer as the curing progresses, and print quality can be improved.

The light-curing polymer is required not only to be water soluble but also to have hydrophilic groups and photoreactive groups (preferable ultraviolet-reactive groups) in its molecules. Some examples of the hydrophilic groups to be contained in the molecules include hydroxyl groups, ethylene glycol chains, propylene glycol chains, etc., and it is preferable for each molecule to contain several hydrophilic groups. Similarly, the photoreactive groups to be contained in the molecules are photopolymerizable functional groups, such as acrylic groups, etc., and it is preferable for each molecule to contain several photoreactive groups. The molecular weight of these light-curing polymers can vary in a wide range as long as the intended effects can be obtained. Accordingly, light-curing polymers having from a low molecular weight, like oligomers, to a high molecular weight of 300,000 or higher can be optionally used.

For reference, these light-curing polymers are commercially available under the following trade names:

Acrylic light-curing oligomers

Trade names "Aronix TO-1321" and "Aronix TO-1343" (both are products of Toagosei Chemical Co.)

Acrylic light-curing polymers

Trade name "NK polymer B-1516S-11" (Shin Nakamura Chemical Co.)

Fig. 9 is a schematic drawing for plainly explaining the change in the water solubility of light-curing polymers before and after polymerization. The light-curing polymer (I) prior to polymerization is soluble in water because it has hydrophilic groups C within its main chain and photoreactive groups D as its branched chains. However, when, after the completion of a printing process, the ink receptive layer that holds an ink image is exposed to light, such as ultraviolet rays (UV), etc., that can induce the polymerization and curing of light-curing polymers, the photoreactive groups (D) crosslink three-dimensionally since there are many of them in one molecule, thereby decreasing the hydrophilic portions; as a consequence, the polymers become insoluble in water (that is, hydrophobic).

In addition to the aforesaid ink receptive element, the present invention also provides a method for implementing printing on this ink receptive element by ink jet methods. Since the ink receptive element used in the method of the present invention is the ink receptive element of the present invention that contains a base material that in

itself does not have ink receptivity, no detailed explanation for this element is given here to avoid repetition.

The following lists some preferable modes of implementing the printing method of the present invention.

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(1) A printing method that implements printing with an aqueous ink by an ink jet method after providing, on the printing surface of an ink receptive element, an ink receptive layer that is transparent and that contains, as the main component, a film-forming polymer formed by the polymerization of a water-soluble monomer having hydrophilic moieties in its molecules, in which printing method the aforesaid ink receptive layer is provided by forming the aforesaid film-forming polymer by the homopolymerization of the aforesaid water-soluble monomer in a film-forming composition containing the aforesaid water-soluble monomer and other essential components and subsequently by applying and drying the obtained resin solution on the base material.

(2) A printing method that implements printing with an aqueous ink by an ink jet method after providing, on the printing surface of an ink receptive element, an ink receptive layer that is transparent and that contains, as the main component, a film-forming polymer formed by polymerization of a water-soluble monomer having hydrophilic moieties in its molecules, in which printing method the aforesaid ink receptive layer is provided by mixing a resin solution containing a homopolymer derived from the aforesaid water-soluble monomer with a

light-curing polymer that is soluble in water and that has hydrophilic groups and photopolymerizable functional groups and subsequently by applying and drying the aforesaid resin solution on the base material, and the aforesaid light-curing polymer is cured by the irradiation of light after the printing process is completed.

(3) A printing method that implements printing with an aqueous ink by an ink jet method after providing, on the printing surface of an ink receptive element, an ink receptive layer that is transparent and that contains, as the main component, a film-forming polymer formed by polymerization of a water-soluble monomer having hydrophilic moieties in its molecules, in which printing method the aforesaid ink receptive layer is provided by forming the aforesaid film-forming polymer by the copolymerization of the aforesaid water-soluble monomer and a water-soluble crosslinking monomer having hydrophilic groups in its molecules in a film-forming composition containing the aforesaid water-soluble monomer, the aforesaid crosslinking monomer, and other essential components and subsequently by applying and drying the /20 obtained resin solution on the base material.

(4) A printing method that implements printing with an aqueous ink by an ink jet method after providing, on the printing surface of an ink receptive element, an ink receptive layer that is transparent and that contains, as the main component, a film-forming polymer formed by polymerization of a water-soluble monomer having hydrophilic moieties in its molecules, in which printing method the aforesaid ink

receptive layer is provided by further mixing a resin solution containing a film-forming polymer derived from the copolymerization of the aforesaid water-soluble monomer and water-soluble crosslinking monomer having hydrophilic groups in its molecules with a light-curing polymer that is soluble in water and that has hydrophilic groups and light-curing functional groups and subsequently by applying and drying the aforesaid resin solution mixture on the base material, and the aforesaid light-curing polymer is cured by the irradiation of light after the printing process is completed.

(5) The printing method stated in the aforesaid item 2 or 4, in which the aforesaid light-curing polymer is comprised of an ultraviolet-curing polymer and is polymerized and cured by the irradiation of ultraviolet rays after the completion of the printing process.

(6) The printing method stated in any one of the aforesaid items 1 through 5, in which the aforesaid ink receptive layer is provided after an undercolor coating is formed on the printing surface of the aforesaid base material.

(7) The printing method stated in the aforesaid item 6, in which, after the aforesaid undercolor coating is formed, its surface is further subjected to a hydrophilicity-imparting treatment.

(8) The printing method stated in the aforesaid item 7, in which the aforesaid hydrophilicity-imparting treatment is an ultraviolet ozone treatment.

(9) The printing method stated in any one of the aforesaid items 1 through 8, in which the aforesaid water-soluble monomer is N,N-dimethyl acrylamide.

The ink jet printing method of the present invention is described in the following, taking as an example a case of implementing printing on a clockface having the same structure as that of the /21 ink receptive element (10) that has been explained in reference to Fig.3. In the explanation presented below, the alphabet in parentheses that is assigned to each process step corresponds to the alphabet in Fig. 10.

(A) Formation of an undercolor coating on the clockface

On one side of a plastic base material (polymer ester sheet) was applied a coating having the color intended for the clockface, thereby forming an undercolor coating (3). In the example illustrated in the figure, a white urethane coating material was applied by electrostatic painting.

(B) Hydrophilicity-imparting treatment for the undercolor coating

A clear coating (4) was further formed on the white undercolor coating (3). The clear coating (4) can be formed by applying, for example, a urethane-based clear coating material and by drying it. Thereafter, the clear coating (4) was subjected to an ultraviolet-ozone treatment, thereby imparting a prescribed level of hydrophilicity to the coating. Here, the evaluation of the obtained hydrophilicity was conducted, as shown in Fig. 11, by measuring

dynamic contact angles (advancing contact angle and receding contact angle) of pure water. For example, the advancing contact angle is measured by dropping pure water (22) on the surface of a test plate (21) with a nozzle (24), as shown in Fig. 11 (I). The pure water (22) expands as indicated with the arrows in the figure as it is supplied. The receding contact angle, as shown in Fig. 11 (II), is measured by suctioning pure water with the nozzle (24) from the large pure-water droplet (22) formed in the aforesaid manner. The water is drawn up as indicated by the arrows in the figure.

With the method of the present invention, the advancing contact angle is preferably  $80^{\circ}$  or smaller, better yet,  $60^{\circ}$  or smaller. The receding contact angle is preferably  $40^{\circ}$  or smaller, better yet,  $30^{\circ}$  or smaller.

Incidentally, these dynamic contact angles become smaller as the duration of the ultraviolet-ozone treatment becomes longer. Fig. 12 attached herein is a graph obtained by plotting the relationship between the duration (in minutes) of the ultraviolet-ozone treatment /22 and the dynamic contact angles ( $^{\circ}$ ). In the figure, Curve I represents the advancing contact angle ( $^{\circ}$ ), and Curve II represents the receding contact angle ( $^{\circ}$ ).

In addition, when implementing an ultraviolet-ozone treatment according to the method of the present invention, it is preferable to keep the fading of the undercolor coating caused by this treatment to



a low level. Usually, the fading is preferably a maximum of 0.5, as expressed in color deference  $\Delta E^* ab$ , which is a common practice in the field of color science. Fig. 13 attached to the present specification is a graph obtained by plotting the relationship between the duration (in minutes) of the ultraviolet-ozone treatment and color deference  $\Delta E^* ab$ . For the specifics of color deference  $\Delta E^* ab$ , please refer to the CIE 1976  $L^*a^*b^*$  color difference formula.

As can be seen from Figs. 12 and 13 presented in the foregoing, with respect to the duration of the ultraviolet-ozone treatment, a treatment of 2 minutes or thereabouts can usually yield the intended effect, although it may vary depending on such factors as the intensity of the treatment, etc., and, from the standpoint of inhibiting color fading, it is desirable to complete the treatment in about 7.5 minutes. Furthermore, the ideal treatment duration is in the range of 3 to 7 minutes.

In the aforesaid process, an ultraviolet-ozone treatment was implemented, but, in place of this, other conventional surface treatments, such as coupling, etching, etc., may be implemented to impart hydrophilicity.

#### (C) Formation of an ink receptive layer

A transparent resin solution containing a film-forming polymer formed by the copolymerization of a water-soluble monomer and a crosslinking monomer having hydrophilic groups in its molecules, a water-soluble light-curing polymer having hydrophilic groups and

light-curing functional groups in its molecules (in this example, an ultraviolet-curing polymer is used), and other essential components was applied to the uppermost layer of the base material (1), that is, to the clear coating (4), and subsequently dried to evaporate the water content contained in the solution. Here, spray coating was employed to apply the resin solution, but other conventional coating /23 methods, including bar coating and printing, such as screen printing, etc., may also be used. This step thus completes the formation of an ink receptive layer (2) and, as a result, ink receptive element (10).

#### (D) Printing by an ink jet method

Full color printing was carried out with an ink jet printer on the transparent ink receptive layer (2) formed in the aforesaid process step to create patterns and dial readings. The ink used here was an aqueous pigment ink. After the ink had permeated into the ink receptive layer (2), the ink receptive element was dried in order to eliminate the water content and humectant component of the ink. This drying can be implemented by using a conventional circulating-type thermostatic chamber, etc. The drying temperature is usually 60 °C or higher, and the drying period is 3 hours or longer. In this manner, a printed ink layer (5) is created.

#### (E) Ultraviolet ray curing process

After the printed ink layer (5) was dried, the layer was exposed to ultraviolet rays (see the arrows) emitted from an ultraviolet curing apparatus, thereby curing the ultraviolet-curing polymer

contained in the resin solution. The curing conditions may vary depending on the absorption wavelength of the employed photopolymerization initiator, but a conventional light source (wavelength: 300 to 450 nm) used for ultraviolet curing can be employed here. By the curing of the ultraviolet-curing polymer, the ink is incorporated into the polymer, thereby forming a printed ink layer (5) that was firmly fixed.

To give a further explanation, the ink receptive element and the printing method of the present invention can employ various types of inks as the recording liquid, but it is preferable to use aqueous inks, particularly those designed to be used for ink jet recording, most of all, aqueous pigment inks.

The aqueous pigment ink used here is comprised of a minimum of a dispersant, water-soluble liquid medium, humectant, pigment, and water, which are commonly used for ink jet recording inks. Each ink /24 component can be selected from those that are conventionally used, and each component is briefly described in the following.

The dispersant is added in order to stably disperse a pigment and thereby to achieve a particle size generally in the range of 50 to 200 nm, and some examples include water-soluble resins and surfactants that have this property and whose hydrophilic moieties are comprised of anionic groups, such as carboxylic acid groups, sulfonic acid groups, phosphoric acid groups, etc., or of nonionic substituents, such as polyoxy alkylene groups, etc. It is preferable for this

dispersant to have a molecular weight of several hundreds to tens of thousands, although it may vary depending on the type of pigment to be dispersed.

The water-soluble liquid medium is used as one of the counter ion species of the anionic dispersant and includes various types of alkanol amines, such as diethanol amine, triethanol amine, etc. Some examples of the counter ion species of the dispersant other than alkanol amines include alkali metals, such as sodium, potassium, etc., and ammonia. Other useful water-soluble liquid mediums include permeability-imparting agents, pH regulators, surface tension regulators, viscosity regulators, and so forth.

The humectant is used for the purpose of stabilizing the dispersion system and adjusting the viscosity, etc., of the dried ink after the evaporation of water, and some examples include ethylene glycols, such as ethylene glycol, polyethylene glycol, etc.; propylene glycols, such as propylene glycol, polypropylene glycol, etc.; diols, such as 1,3-butane diol, 1,3-propane diol, etc.; and pyrrolidones. These humectants may be used singly, but, when considering the stability of the dispersion system and the like, it is desirable to use a humectant mixture system prepared by mixing several kinds of humectants. /25

The pigment can be selected from a wide range of known pigments that can meet such requirements as the required color, compatibility with the dispersant, and so forth, and should be used in a proper

quantity. For example, typical examples of black pigments are carbon blacks, typical examples of blue pigments are phthalocyanine pigments, typical examples of red pigments are quinacridone pigments, and typical examples of yellow pigments are azo pigments.

The ink components described in the foregoing, according to such factors as the ink jet printer to be used, the type of printed product, and so forth, can be formed into a solution having an appropriate viscosity by mixing and kneading them in optional proportions.

The ink receptive element and printing method of the present invention can each be advantageously utilized for preparing various types of printed products. Ideal printed products include, but are not limited to, clockfaces, the dial plates of various kinds of meters installed in automobiles and measuring instruments, and so forth.

#### Working Examples

In the following, the present invention is further described, referring to working examples. It should, however, be borne in mind that the present invention is not limited to or by the following working examples.

##### Example 1

A reactor was filled with 80 parts by weight of water and subjected to nitrogen replacement. Thereafter, 10 parts by weight of acrylamide (as the monomer having amide groups) and 0.01 part by weight of ammonium persulfate (as the polymerization initiator) were added to the reactor, and the reaction system was polymerized at 60 °C

for 45 minutes. After the polymerization reaction was completed, a /26  
solution obtained by dissolving 0.7 part by weight of methylene  
bisacrylamide (as the crosslinking monomer) in dimethyl formamide was  
added to the reaction system. Two hours later, 0.01 part by weight of  
ammonium persulfate was further added to the reaction system, and the  
polymerization was continued for 12 hours. After the continued  
polymerization reaction was completed, 2.5 parts by weight of acrylic  
light-curing polymer (product name "NK polymer B-1516S-11") and 0.6  
part by weight of a photopolymerization initiator (product name  
"Irugacure 1173") were added to the reaction system. In this manner, a  
transparent resin solution to be used for the formation of an ink  
receptive layer was obtained.

#### Example 2

The same procedures as in Example 1 above was repeated, but, in  
this example, acrylamide was replaced by the same amount of N, N-  
dimethyl acrylamide, methylene bisacrylamide was replaced by the same  
amount of polyethylene glycol #600 diacrylate, and, as the acrylic  
light-curing polymer, "Aronix TO-1343" (a product name) was used in  
place of "NK Polymer B-1516S-11" (a product name) in the same amount.  
In this manner, a transparent resin solution to be used for the  
formation of an ink receptive layer was obtained.

#### Example 3

The same procedures as in Example 1 above was repeated, but, in  
this example, acrylamide was replaced by the same amount of N, N-

dimethyl acrylamide, and methylene bisacrylamide was replaced by the same amount of polyethylene glycol #600 diacrylate. In this manner, a transparent resin solution to be used for the formation of an ink receptive layer was obtained.

#### Example 4

The same procedures as in Example 1 above was repeated, but, in this example, acrylamide was replaced by the same amount of acryloyl morpholine, methylene bisacrylamide was replaced by the same amount of 2,2-bis[4-(methacryloxy polyethoxy) phenyl] propane, and, as the acrylic light-curing polymer, "Aronix TO-1343" (a product name) was used in place of "NK Polymer B-1516S-11" (a product name) in the same amount. In this manner, a transparent resin solution to be used for the formation of an ink receptive layer was obtained. /27

#### Comparative Example 1

A reactor was filled with 80 parts by weight of water and subjected to nitrogen replacement. Thereafter, 7 parts by weight of vinyl pyrrolidone [sic], 3 parts by weight of N,N-dimethyl acrylamide, and 0.3 part by weight of 2,2'-azobis (2-amidinopropane) dihydrochloride (as the polymerization initiator) were added to the reactor, and the reaction system was polymerized at 70 °C for 45 minutes. In this manner, a transparent resin solution was obtained.

#### Comparative Example 2

Ten parts by weight of polyvinyl alcohol (saponification degree: 100%, polymerization degree: 1700) was dissolved in 90 parts by weight of water, thereby obtaining a transparent resin solution.

#### Comparative Example 3

A reactor was filled with 80 parts by weight of water and subjected to nitrogen replacement. Thereafter, 10 parts by weight of acrylamide and 0.01 part by weight of ammonium persulfate (as the polymerization initiator) were added to the reactor, and the reaction system was polymerized at 60 °C for 45 minutes. In this manner, a transparent resin solution was obtained.

#### Comparative Example 4

A reactor was filled with 80 parts by weight of water and subjected to nitrogen replacement. Thereafter, 10 parts by weight of N,N-dimethyl acrylamide and 0.01 part by weight of ammonium persulfate (as the polymerization initiator) were added to the reactor, and the reaction system was polymerized at 60 °C for 45 minutes. After the polymerization reaction was completed, a solution obtained by dissolving 0.7 part by weight of methylene bisacrylamide (as the crosslinking monomer) in dimethyl formamide was added to the reaction system. Two hours later, 0.01 part by weight of ammonium persulfate was further added to the reaction system, and the polymerization was continued for 12 hours. In this manner, a transparent resin solution was obtained.



### Example 5

The resin solution formulated in each of Examples 1 through 4 and Comparative Examples 1 through 4 was applied to a white dial plate with a bar coater and dried at 60 °C for 3 hours. The obtained ink receptive layer was tested according to the following procedures. /28

#### (1) Film property test

The obtained white dial plate was stored and dried for 1 week at normal temperature (23 to 25 °C) and low humidity (20% or less). The dried ink receptive layer was visually observed, and, if it did not have any defect, "O (pass)" was assigned to the test piece, while "X (fail)" was assigned to it if the film showed peeling or cracking.

#### (2) Printing test

Full color printing was carried out with an ink jet printer that was equipped with aqueous pigment inks, thus printing a pattern on the white dial plate. The printed condition was visually observed, and, if the print did not have any defect, "O (pass)" was assigned to the test piece, while "X (fail)" was assigned to it if the print showed bleeding or color mixing.

#### (3) Adhesion test

The adhesion strength of the ink receptive layer to the white dial plate was evaluated by a cross-cut adhesion test (in accordance with JIS D0202 8. 12). A grid was formed on the ink receptive layer with a cutter, and an adhesive tape was attached to the layer. After the elapse of a given time period, the adhesive tape was peeled off,

and the presence or absence of the peeling of the ink receptive layer was examined. "O (pass)" was assigned to the test piece when no peeling occurred, while "X (fail)" was assigned to the test piece when peeling took place.

(4) Light resistance test

The white dial plate was mounted vertically in a sunshine weatherometer (light source: a carbon arc lamp) and left standing in a dry condition (normal humidity) or wet condition (saturated steam) for 100 hours. "O (pass)" was assigned to the test piece when no fading was observed in the pattern on the white dial plate after it was left standing under the aforesaid condition, and "X (fail)" was assigned to the test piece when fading was observed.

(5) Humidity resistance test

The white dial plate was mounted horizontally or vertically in /29 an environmental tester (a constant temperature/humidity chamber) and left standing at 40 °C /90% RH (relative humidity) for 100 hours. "O (pass)" was assigned to the test piece when no change was observed in the pattern on the white dial plate after it was left standing under the aforesaid conditions, and "X (fail)" was assigned to the test piece when changes, such as running, etc., were observed.

Table 1 below summarizes the results of the aforesaid tests (1) through (5).

TABLE 1

Example No.	Film Property Test	Printing Test	Adhesion Test	Light Resistance Test		Humidity Resistance Test	
				Dry	Wet	Horizontal	Vertical
Ex. 1	O	O	O	O	O	O	O
Ex. 2	O	O	O	O	O	O	O
Ex. 3	O	O	O	O	O	O	O
Ex. 4	O	O	O	O	O	O	O
Comp. Ex. 1	X	O	X	-	-	-	-
Comp. Ex. 2	O	X	O	-	-	-	-
Comp. Ex. 3	O	O	O	O	X	X	X
Comp. Ex. 4	O	O	O	O	X	O	X

Note) The symbol "-" means "not measurable."

#### Industrial Field of Application

As explained in the foregoing, the present invention can provide an ink receptive element that can be used for clockfaces, the dial plates, operating panels, etc., of measuring instruments, the dial plates, etc., of speedometers for automobiles, motorcycles, etc., and also can provide an ink jet printing method that uses the ink receptive element of the present invention.

#### CLAIMS

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1. An ink receptive element containing a base material that in itself does not have ink receptivity, the aforesaid ink receptive element having, on its printing surface, a transparent ink receptive layer that contains, as the main component, a film-forming polymer prepared by the polymerization of a monomer that is soluble in water and that has hydrophilic moieties in its molecules.

2. The ink receptive element stated in Claim 1, which, in addition to the aforesaid film forming polymer, further contains a